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SEA LEVEL

By HOWARD S. RAPPLEYE*

Going back, in our imagination, to the early stages of civilization on this earth, we may find a man having established his home somewhere in the flat country just back of a gently-sloping sand beach. He must have done this for reasons which seemed good to him, and if we pursue our inquiry further, we would suggest that they may have been about as follows:

(1) The flat fertile country of the lowlands would make it easier for him to grow the food he needed than if he cultivated the steeply-sloping hillsides farther inland.

(2) He would be near an additional source of food supply, to be taken from the sea.

(3) He would have more ready means of communication with his like-minded fellows by means of water transportation.

Our remote ancestor, then, has established his home on the coast and it is not long before he has learned from experience that if he lands his dugout on the beach when the water's edge is far out from his door and does not pull his canoe well up on the beach, it will probably float away within a matter of relatively few hours. Conversely, if he lands when the water's edge is in close to his door he will, if he wishes to set sail again when the water has receded, have to drag his boat a long way down the beach before it can be floated.

This state of affairs is *forced* on his attention and he eventually notices that there is a somewhat rhythmic quality to the behavior of the water's edge with relation to the tree line back of the beach.

He talks the matter over with his family and perhaps with some of his neighbors. They agree that there is a tendency toward a repetitive cycle in the events concerning the behavior of the water's edge. Someone, perhaps one of his great-grandchildren, eventually tumbles to the fact that this cycle of events comes fairly close to repeating itself every time the moon goes through its changes from new moon to half moon, full moon, half moon and back to new moon again. Once this idea has dawned on these people and they seek to prove that it is a fact, the study of the tides has begun.

As it is not within the scope of this discussion to trace the history of tidal in-

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vestigations from the beginning up to the present time, the best we can do is to start out with the knowledge accumulated through the ages and to try and see of what use this knowledge may be to us and what practical results may be obtained from all the investigations that have been completed, as well as from those that are still going on. We may even try to predict what may happen in the future to that much talked-about but too little understood surface which we glibly call *sea level.*

Those who dwell along the shores of the oceans and tidal estuaries will readily admit a keen interest in the tides and there need be no argument about it. That Mrs. John Doe, of Peoria, Illinois, has any direct interest in matters pertaining to the tides is less obvious, though a little reflection may serve to prove that this is so. For example: the accuracy and quality of nautical charts is largely dependent upon an accurate knowledge of the tides and their behavior; good nautical charts, properly used, tend to reduce greatly the dangers to shipping; increased safety for shipping tends to reduce marine insurance rates; and these reduced insurance rates decrease the ultimate cost to the consumer of goods brought from foreign ports. I believe that it will now be agreed that Mrs. Doe pays less for her Italian-lace table cloth or her Chinese-silk kimono than she would have had to pay if a lot of observation and analysis had not gone into the study of the tides.

Since it is obviously much easier to understand a Chinese if you know what he is talking about, it may be well, at this point, to define sea level and explain briefly how it is determined.

Mean sea level is defined, by those best qualified to know just what it really is, as "the average height of the surface of the sea, all stages of the tide considered." This definition implies measurement of the height of the water surface with reference to some point or index. Since mean sea level is determined by measurement, the resulting data must perforce apply to the surface of the sea at the place where the observations were made. Further, the length of the series of observations has a great effect on the accuracy of the resulting numerical value. For these reasons, it is quite customary, in defining mean sea level, to state the length of the series of observations on which the numerical value is based. Thus, we might say that mean sea level at Fort Hamilton, N. Y., based on continuous observations for 38 years, from 1893 to 1930, is 8.98 feet below bench mark 20.

The modern method of determining mean sea level is to establish a tide gauge and operate it for a considerable period of time. This gauge is simply an installation which produces a continuous graphic record of the rise and fall of the surface of the water, with an appropriate datum line running the length of the continuous curve and suitable time marks spaced at intervals of one hour along the curve.

FIGURE 1 shows a schematic diagram of the installation and you will note that the recording instrument is mounted on the table or bench inside the tide-gauge house, and that the bench and instrument are placed over the float well so that a vertical wire leading from the float to the recording instrument will hang in the center of the well and keep the float from dragging on the sides of the well. Two counter-weights show in the diagram. One is in a bight in the wire leading from the float over a spool on the gauge and through suitable pulleys to the counterweight and then to an anchorage in the roof. The other furnishes the power for the take-up spool to keep the paper wound up properly and to relieve some strain on the clock that controls the feeding of the paper over the main roller, along an element of which the recording pencil moves back and forth in trac-

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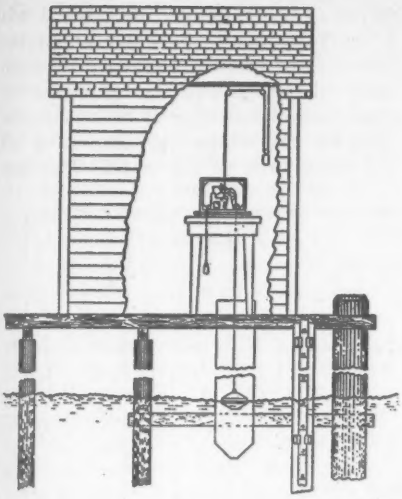

FIGURE 1.

ing the curve of the rise and fall of the float in the well.

In a close-up of the recording gauge, we would notice two clocks, one causing the paper to feed over the main roller at the rate of one inch per hour and the other producing the time marks at hourly intervals on the curve traced by the recording pencil. A fixed pencil traces a datum line the length of the curve as the paper passes under it.

The curve is traced by means of a pencil which is made to move along an element of the main roller by means of a spirally-grooved shaft which in turn is directly connected to the spool over which the wire passes in leading from the float to the counter-weight. This counter-weight keeps the wire taut at all times and prevents slippage on the spool, which is grooved and carries several turns of wire at one time to insure sufficient friction between the wire and the spool.

The tide staff is simply a long board, graduated in feet and tenths from the bottom upward, and is mounted in a vertical position on some suitable structure so that its zero is well below the lowest expectable low stage of the tide at that station. This staff is read daily at the time the gauge is inspected and serviced, and the staff reading is noted directly on the curve at the point indicated by a manually-made jog, similar to an hourly time mark, put in the curve at the time the staff reading was taken. These staff readings serve to define the height of the datum line on the curve with respect to the zero of the staff gauge.

At suitable intervals of time, the staff is connected, by means of spirit leveling, with the bench marks on shore. These are set at suitable locations on shore and near the tide station. They serve to furnish the permanent points to which the observations are referred.

Should the staff be displaced by a storm or some other accident, a new one can be installed and the observations kept continuous by leveling to the new staff from the bench marks and determining accurately the height difference between the zero of the original staff and the zero of the new one. Properly handled, the series of observations at a station may be kept on a consistent basis, even though the staff may have been dislodged once or even many times during a long series of observations.

At many places, the tide is a semi-daily phenomenon, the surface of the water going up and down again twice in one day. We may look at the tide curve for New York, say, for the month of June, 1934. The actual curve traced by the recording gauge is 24 inches long for each day and cannot be more than about 12 inches high, even when the full available height limits are utilized. Suitable reduction mechanisms will reduce even a 30-foot tide to curve height of a foot or less. Thus, it will be seen that the curve in question has been greatly compressed in the matter of time. Our illustrations are usually made up in this way to conserve valuable space.

While the semi-daily tide is probably best known, especially to the layman, this comes about, not because it is the most prevalent type of tide but because it happens to be the type of tide which occurs at most places on the shores of the Atlantic Ocean and has been observed at more stations. The mixed tide is certainly more prevalent along the shores of the Pacific Ocean than the semi-daily tide and, therefore, is probably the most widespread type of tide when we consider the whole aggregation of all the shore lines of the world. The true

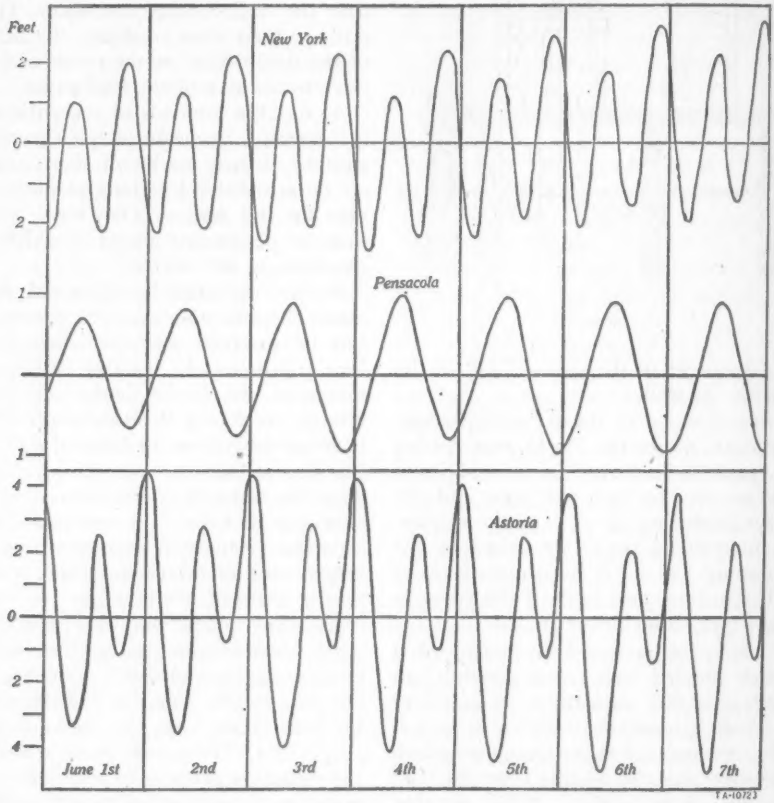

FIGURE 2.

daily tide is, without doubt, less frequently encountered than either the semi-daily or mixed tides.

If we look at the tide curve for Seattle, Washington, also for the month of June, 1934, we can see that the tide at Seattle is quite different from that in New York and, also, that the character of the tide changes quite materially during the month.

FIGURE 2 shows a week of record for each of three stations, New York, Pensacola, and Astoria. These, in order from top to bottom, represent typical examples of the semi-daily, daily, and mixed tides for purposes of comparison.

It will be noticed that, in the schematic diagram of the tide-gauge installation (FIGURE 1), the bottom of the float well is tapered to a point. This is to indicate that the bottom of the well is constricted to a small opening to damp out wave action of short period and to produce a smooth curve free from the violent short-period oscillations that would be produced by severe storms. This not only serves to smooth the curve and permit more accuracy in scaling the hourly heights from the curve, but saves wear and tear on the gauge itself. If the bottom of the float well were wide open, wave action might cause the float to rise so rapidly that the counter-weight could not keep sufficient tension on the wire, and the wire would come off the grooved wheel at the end of the spirally-grooved shaft. The result would be that the gauge would be out of action until again serviced and there would be a gap in the record. Further, the small opening in the bottom of the float well is *actually* at the lower extremity of the conical bottom to permit finding the opening quickly when it is necessary to pass a cleaning rod through it to dispose of barnacles or other matter that may have partially clogged the opening.

By means of the recorded staff readings, the height of the datum line on the curve is determined and the hourly heights are scaled and tabulated. These are averaged to produce the final result. It may be interesting to note that the 38 years of observations at Fort Hamilton, to which we previously referred, involved at least 456 marigrams, each at least 60 feet long, and the scaling and averaging of over 330,000 hourly heights.

The averaging of the 24 hourly heights for a single day results in a daily mean sea level. For the rather obvious reasons of wind, variation of barometric pressure, etc., this will vary considerably from day to day. In a study of actual records of the values of daily mean sea level at various locations, it will be noticed that the daily mean sea levels vary considerably from day to day, in places as much as a foot or more between two consecutive days. Obviously, if we want an *accurate* determination of mean sea level we will have to extend our observations over a longer period of time.

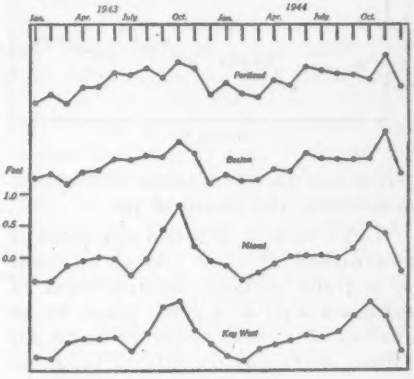

FIGURE 3.

The averaging of the scaled hourly heights for a month results in a monthly mean sea level. Certain typical observations of monthly mean sea levels are show in FIGURE 3. Here, we have shown monthly mean sea levels for a two-year period for each of four Atlantic Coast stations: Portland, Maine; Boston, Massachusetts; and Miami and Key West, Florida. A close examination of FIGURE 3 will show two things:

(1) There is still a considerable variation from month to month, in some places approximately half a foot.

(2) There is obviously a seasonal effect.

That this seasonal effect is not merely an apparent condition but actually exists, is shown by FIGURE 4. Here, we have *daily* mean sea levels for the months of January and July, 1939, at Atlantic City, N. J., and it is rather obvious that during the month of January the behavior of the

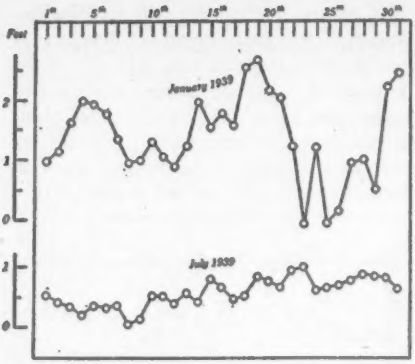

FIGURE 4.

tide at that place was much more erratic than during the month of July.

It now looks as if a two-year series of observations were not enough to result in a really accurate determination of mean sea level at a given place. In an analysis of annual mean sea levels for various stations on the Atlantic Coast, we find that the variation from year to year is much less than the variation from day to day or month to month, but that even there considerable variation (up to as much as 0.2 ft.) occurs between successive annual means at these stations.

Theoretically, a series of sea levels taken over some twenty years should produce a resulting mean sea level which is accurate to within a hundredth of a foot, and, actually, in most localities, this is true. However, such a series, when examined closely, will show an apparent upward trend with the passage of time at various stations on all coasts of the United States, so that it looks as if it were still necessary to keep comparison stations going continually at many points along the coasts in order to study the long-term behavior of this reference surface, which is apparently continually developing some new whim or twist. But more of this later.

It will be evident from the foregoing statements that the task of making a really accurate determination of mean sea level at a given station is a long and tedious process. It requires the establishment and maintenance of an automatic water-level recording instrument, daily inspection, recording the daily reading on the staff and the time at which the reading was taken, the periodic cleaning of the float well, occasional replacement of the staff or even of the recording instrument, and periodic levelings to the staff from the bench marks on shore. In the office, there are thousands of hourly heights to be scaled from the marigrams each year, monthly means to compute and record, annual means to be taken, and all this costs money. The cost of a really accurate determination of mean sea level is such that it is readily understood why there are not more long-period tide stations in operation on the coasts of this and other countries.

Mean sea level having been determined at a station, we are now at liberty to consider some of the associated datum planes or surfaces that are intimately connected with the principal plane.

Half-tide level is, as might be expected, the average of all the high and low points on the tide curve. This plane or surface of reference is usually the one that is computed and published in lieu of mean sea level at a station where only a short series of observations is available. It is a fairly close approximation to mean sea level, but may vary from the value of mean sea level, which might be computed from the same series of observations, by as much as 0.2 ft. or more in some cases.

The navigator is interested in knowing the least depth of water over shoals and bars. For this reason, the datum to which soundings are referred in the preparation of nautical charts is usually a low-water datum. On the Atlantic Coast, and in other places where the daily or semi-daily tides are the rule, the plane of reference to which soundings on charts

are reduced is mean low water and, mathematically, this is the average of all the low waters during the period of the observations.

In localities where the mixed tide occurs, the plane of reference for soundings is usually mean lower low water, since there is usually considerable difference between the heights of the two low waters which occur in a single tidal day at such stations.

The engineer who is interested in the drainage of wet lands near the coast or in disposing of storm water in a coast city is interested not in a low-water datum but in a high-water datum, since the effectiveness of his drainage or disposal system is decreased by high water in the bay or stream into which he is running his unwanted water. Accordingly, the plane of mean high water is used in the vicinity of stations having semi-daily or daily tides and the plane of mean higher high water is the one in which interest is centered in case the mixed tide is encountered in connection with a drainage problem.

Further, the extreme tides are of interest in connection with navigation when unusual conditions are encountered and in drainage when periods of extreme high water are apt to have a serious effect on the drainage system or even cause a backing up of unwanted water to make a flood. Accordingly, the extreme high and low tides are usually noted and published along with the rest of the information concerning a particular tide station.

FIGURE 5 is of interest in that it shows what can happen to a tide curve at a given station when a really unusual set of conditions exists for a more or less short period of time. This curve shows the fluctuations of the water surface at Honolulu for the four days, February 2-5, 1923, and reflects the effect of the earthquake which occurred on February 3, 1923, off the coast of Kamchatka, about 2500 miles from Honolulu. The first sea wave arrived at Honolulu about six hours after the earth quake, and the subsequent fluctuations of the water surface were of considerably greater magnitude in a matter of minutes than the normal mean range of the tide at that place.

Lastly, in connection with the published data, the descriptions of the bench

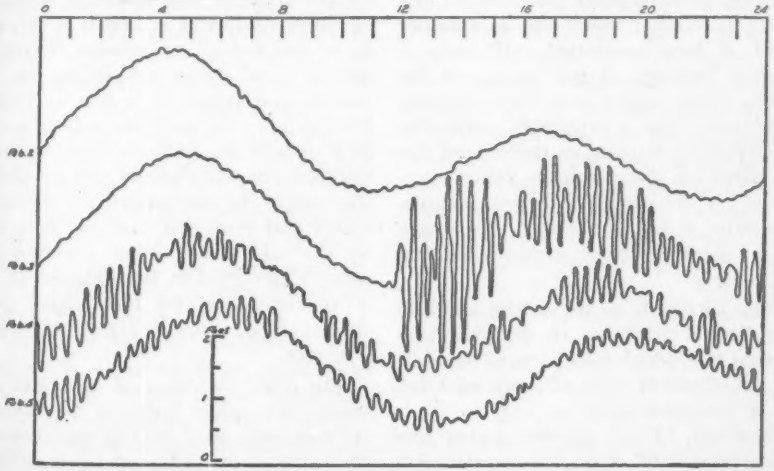

FIGURE 5.

marks to which the tide staff is referenced are listed with their elevations given above one of the reference planes at the station, usually one of the low-water planes. The elevations can be transferred from one datum plane to another by simple arithmetic, as the differences between the various planes may be obtained by simple subtraction.

In all of the preceding discussion, we have tacitly assumed that everything is working smoothly, that the shore line is stable, the plane of mean sea level once accurately determined remains fixed, and we have not made much mention of the effects of wind, currents, variations in barometric pressure, fresh water effect in tidal streams, channel effects when stations are located in bays or harbors having narrow entrances, stability of the land, or variations in the mean level of the sea itself.

Obviously, wind action is most noticeable in form of waves breaking on the shore. We have already discussed the matter of avoiding this difficulty by constricting the opening in the bottom of the float well. Another problem arises in connection with the known fact that a constant onshore wind will tend to pile water up on the beach and an offshore wind, if long continued, will cause a distinct lowering of the surface of the water. Even under less than extreme conditions this is especially noticeable in bays with narrow entrances and has its effect on the resulting values computed for the various reference planes, especially at stations where only a relatively short series of observations has been made.

The direction of prevailing wind in the United States is, in general, from west to east, which may account partially for the observed "tilt" of mean sea level, to be discussed later.

Naturally, a tide station located considerably inland from the mouth of a tidal stream is bound to be under the influence of the fresh water coming down the stream. Floods may raise the water level by many feet for short periods of time and, as these extreme high waters are all included in computing the final values of the reference planes, they have their effect and tend to make the reference planes, at stations not on the open coast, stand higher than the corresponding planes at the open-coast stations. Also, fresh water is of different density than salt water. This causes a slight added complication when dealing with a station at which a large amount of fresh water passes the gauge.

At this point, it would be reasonable to ask why, if so much trouble is encountered with tide stations that are not on the open coast, not all stations are built outside, where they will not encounter all of the difficulties?

There are two principal answers to that question:

(1) A tide station on the open coast is very costly to construct and difficult to maintain, except perhaps where one is lucky enough to find a place like Atlantic City where piers actually extend out into the open ocean, for the station usually requires that a special structure be built to support it, and it is very apt to be put out of commission by storms which require the rebuilding of the station and result in a loss of record. Furthermore, in order to reduce soundings in bays and tidal streams, we need the local reference planes and not planes that might be determined on the open coasts and projected into the bays and up the tidal streams. For example, the mean water level at Washington, D. C., is approximately 0.4 foot higher than the mean sea level at Hampton Roads, Virginia.

(2) Also, the *times* of high and low waters are quite different at different stations and, in reducing soundings to the adopted reference plane, simultaneous tide-gauge readings are needed at a

location close by the area which is being sounded by the hydrographic party.

We have touched upon the needs of the navigator and discussed the reduction of soundings to the adopted reference plane. This provides the mariner with a knowledge of what is apt to be least water over the bars and shoals which he will encounter. However, especially at harbor entrances, a deep-draft vessel may be required to wait for high tide to successfully cross the bar. This requires that the navigator be provided with the times and heights of high and low waters at many places in advance of his sailing. The tide tables published by the Coast and Geodetic Survey, and by other similar organizations in other countries, give the mariner this information based on predictions of the time and height of high and low water at many stations.

The observations which we have been discussing are also subjected to a mathematical process known as harmonic analysis, and certain constants are determined for each long-period station. From these constants can be worked out the predicted tides for future use. This, in the old days, was a really laborious process, but by the use of the tide-predicting machine which was developed in the offices and shops of our Bureau the whole process becomes a mechanical one.

The necessary constants for a station can be set in the machine and checked and the predictions for a year for that station completed in one day. Usually, the predictions are made two or three years in advance so that the information may be printed and distributed in advance of the time it will be needed by the mariner. It is rather remarkable what accuracy is obtainable in these predicted values. Unless storm conditions are encountered, the predicted values are usually checked, when the tide observations are actually made at the station, within a few minutes in the *times* of high

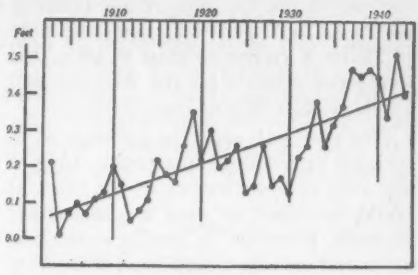

FIGURE 6.

and low waters and within a tenth of a foot or so in the *heights* of high and low waters.

For the stations on the Atlantic Coast, there was an evident upward trend of the mean sea level surface on the staff at each station. FIGURE 6 shows the annual mean sea levels at Baltimore, Maryland, from 1903 to 1943, and there has been drawn through the irregular curve representing these annual mean sea levels a straight line which was located on the other curve by the method of least squares. This, *if* we assume that sea level has been rising at a uniform rate during the forty-year period, gives the rate of rise, which appears to be something of the order of size of 0.01 ft. per year.

FIGURE 7 shows the result of treating the same basic data by the method of the moving mean. With this treatment, it appears that the rise has not been uniform, as was *assumed* in the preparation of FIGURE 6, but slow during the period prior to about 1928 and much more rapid from then on to the end of the series in 1943. The greatest slope of the curve shows that, during the last fifteen years,

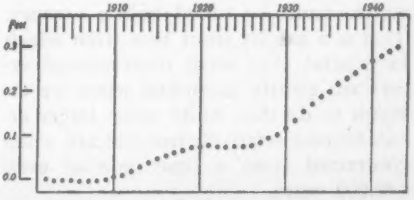

FIGURE 7.

the rate of rise has been of the order of size of 0.015 ft. per year. The same state of affairs is known to exist at all of the long-series stations on the Atlantic and Pacific Coasts of the United States.

One may well raise the question as to whether or not this represents a rising of sea level or a settlement of the land. It could be either, or even a combination of both. However, it hardly seems possible that so large a land mass as the continental United States could be sinking so uniformly as to produce the observed changes in the relationship between the tide staff and mean sea level at so many different tide stations so widely scattered along the coasts of this country. Furthermore, it is a known fact that the glaciers of the world have been receding for a considerable period of time. The melting back of the glaciers and a shrinking of the boundaries of the ice caps at the poles would certainly have some effect on the elevation of mean sea level throughout the world. Also, the soil conservationists have been crying for some time for greater efforts in holding moisture in the soil. Water tables have lowered seriously in many places. All of this is in the right direction to account, partially at least, for the apparent rise in sea level.

The geologist has a habit of measuring time in units of hundreds of thousands or perhaps even millions of years, and finds evidences of very great changes in the level of the sea, the land, or both. As geodesists and tidal specialists, we have to base our conclusions, at present at least, on data which, except in very few instances, barely cover a period which could be called half a century. This is a mighty short base from which to expand. Any small observational errors are greatly magnified when we attempt to do this, while much larger errors become relatively insignificant when contracted from a time base of even 100,000 years.

The fundamental vertical control net of the United States is a very tight network of lines of first-order leveling throughout the country. These have been run in such a way that they form closed loops of spirit leveling and a considerable number of spirit-level connections between the tide bench marks on which our connections to mean sea level are made. An idea of the accuracy of the leveling may be given by stating the fact that, when the combined level nets of the United States and Canada were adjusted simultaneously in 1929, the *average* rate of correction which it was necessary to apply to the approximately 65,000 miles of leveling included in the adjustment was of the order of size of less than 0.2 mm. per kilometer.

It has long been known that geodetic leveling between tide stations did not always produce checks that were within the expected limits of accuracy of the leveling and/or the accuracy of the tide observations. For example, accurate spirit leveling across the Isthmus of Panama shows that mean sea level on the Pacific side is about 3/4 ft. higher than mean sea level on the Atlantic side. The same state of affairs has been observed between the north and south coasts of France, the mean level of the sea on the north coast standing higher than that on the south coast by an amount comparable with the findings in other parts of the world. The same phenomenon was also observed on the coasts of England and Wales, where very accurate spirit leveling failed to agree with the tide observations by amounts considerably greater than could be ascribed to errors in the leveling, errors in the tide observations, or both combined.

In 1927, a special adjustment of the leveling net of the United States was made to test this condition for the coasts of the United States. The problem was attacked by adjusting all of the land circuits of leveling to consistency with-

out holding *any* sea-level connections. Then, with an observed difference from Galveston to the nearest junction in the net, and observed differences from other junctions nearest to the tide stations down to the tidal connections, we were able to arrive at spirit-level elevations for the observed planes of mean sea level at the various tide stations, all based on the observed mean sea level at Galveston. The results showed that, apparently, the sea-level surface, as defined by the observed planes of mean sea level at the various tide stations, had a marked tendency to stand higher to the northward along both the Atlantic and Pacific coasts and higher to the westward along the Gulf Coast. As might be expected from the upward trend westward along the Gulf Coast, the average for the Pacific was considerably higher than that for the Atlantic Coast.

In 1929, a further study was made by making a similar investigation of the combined level nets of the United States and Canada. The results further confirmed the findings of the 1927 Special Adjustment and are shown in TABLE 1, in which the 0.00 difference has been shifted to St. Augustine, simply to rid the table of negative values. The differences between the stations remain the same.

The upward trend along the east and west coasts is quite evident and extends on north along the coasts of the Dominion of Canada. The upward trend along the Gulf Coast to the westward is also evident and the fact that the average height of mean sea level on the Pacific side is considerably higher than the average for the Atlantic stations is also quite plainly shown. As to *why* this condition exists, there are several factors which may be considered. The general direction of prevailing winds may partially account for it. Differences in salinity, variations in average barometric pressure, configuration of the coast lines and, perhaps, ocean currents may each contribute their share to the sum total of this variation of the mean-sea-level surface from its theoretical form. Errors of observation in both the leveling and in the tidal observations are present, of course, but these alone are hardly sufficient to account for the observed differences.

TABLE 1

RELATIVE ELEVATIONS OF MEAN SEA LEVEL DETERMINED BY THE 1929 SPECIAL ADJUSTMENT

Station	*Meters*
Galveston	0.27
Biloxi	0.18
Pensacola	0.22
Cedar Keys	0.19
St. Augustine	0.00
Fernandina	0.07
Brunswick	0.11
Norfolk	0.13
Old Point Comfort	0.01
Cape May	0.29
Atlantic City	0.30
Annapolis	0.10
Baltimore	0.09
Perth Amboy	0.28
Fort Hamilton	0.30
Boston	0.34
Portland	0.39
Yarmouth	0.32
Halifax	0.35
Father Point	0.47
San Diego	0.60
San Pedro	0.58
San Francisco	0.61
Fort Stevens	0.86
Anacortes	0.72
Seattle	0.75
Vancouver	0.77
Prince Rupert	0.85

Such is the present state of affairs, as far as it is possible to picture it in this brief space. We have a host of unsolved problems yet to be worked on. Nature has been keeping at least a crude record of what has been happening for hundreds of thousands of years, for geologists to read. Obviously, the tidal and geodetic observations, which have been carried on intensively in this country for perhaps not more than a half century, give a more detailed picture and permit more accurate quantitative deductions than nature's geologic record. However, the size of the quantities we are trying to measure are dangerously close to the limits of error of the observations themselves, when the short time base on which we now have to work is considered. Given a chance to make accurate tidal and geodetic observations for even *one* thousand years, we shall be able to show results of considerable significance. In the meantime, the geologist will have to make the best of what little we can offer to prove or disprove the interpretations which he makes of nature's record, written in the geolocial structure of the earth.

SECTION OF BIOLOGY

RELATIONSHIPS OF NONHEMOLYTIC AND VIRIDANS STREPTOCOCCI IN MAN

By GEORGE H. CHAPMAN*

In cultures made by the average bacteriologist, a minute colony on a blood agar plate is taken as a streptococcus and any discoloration of the surrounding medium is supposed to differentiate it into "species," although slight differences in ingredients of the culture medium have a profound effect on the blood-altering and other properties of streptococci. Consequently, few reports on the presence of this organism have any significance. I shall try to show how it is possible not only to isolate streptococci more frequently and more surely, but to determine the significance of α- and γ-hemolytic types in man with a high degree of precision. Work with β-hemolytic types presents an entirely different problem.

This report discusses the results of 23 years of intensive study and omits experimental data, much of which has been published elsewhere.[1-30]

Methods. If an apparently pure culture of streptococci is plated so as to give discrete colonies and they are tested separately for different properties, they will be found to constitute a kaleidoscopic mixture. Some may ferment a particular carbohydrate, some may be toxigenic, etc. Moreover, if a toxigenic colony is replated, the daughter colonies, likewise, may vary in toxigenicity. Toxigenic streptococci are particularly significant in man because they are associated with tissue damage and because their cultures make decidedly better antigens than do those of non-toxigenic streptococci. Therefore, precise measurement of the toxigenicity of streptococcal cultures assumes considerable significance. To accomplish this successfully, any degeneration of toxigenicity must be reduced to a minimum. A considerable reduction can be achieved by using brain heart infusion (Difco) for subculture, inoculating it with a single colony to secure a pure undissociated culture rapidly, harvesting the culture in the late logarithmic phase, and transferring it immediately to the refrigerator and testing it promptly. Under these conditions, the proportion of toxigenic colonies in a culture is an accurate measure of its degree of toxigenicity. In general, this proportion of toxigenic colonies in a culture from the body is directly related to the *severity* of intoxication in the tissue from which the culture was obtained. The total number of streptococci in that part is an indicator of the *amount* of toxic material it contains. Such toxigenic organisms probably grow better in the body than do non-toxigenic ones, because usually the higher the total number of streptococci in a particular tissue the greater the proportion of toxigenic colonies.

In acute infections or acute exaccerbations of chronic infections, the organisms become more "virulent" (invasive), but there is little change in toxigenicity.

These toxins can damage or sensitize any tissue in the body. The more intense the toxigenic power of the organisms, the greater the tissue damage. When the damage is severe enough to affect local resistance materially, non-pathogenic bacteria can then invade the tissues. Hence, coliforms and other non-pathogenic bacteria are found in habitats foreign to them usually when the streptococci in that part are highly toxic.[5]

Accurate determination of the total number and proportion of toxigenic colonies in a sample depends on (1) satis-

* Clinical Research Laboratory, New York, N. Y. Aided by grants from the Ophthalmogical Foundation, Inc. This paper, illustrated by lantern slides, was presented at the meeting of the Section on November 10, 1947.

factory quantitative isolation methods and (2) satisfactory methods for testing the toxigenicity of the organisms isolated.

The isolation method used is as follows: Spread 0.01 ml. of appropriate undiluted or serial decimal dilutions of the sample on the surface of a plate of tellurite streptococcus medium, which has the following composition: Proteose peptone No. 3, 5 grams; proteose peptone, Difco, 5 grams; Bacto-tryptose, 10 grams; Bacto-agar, 15 grams; saccharose, 50 grams; K_2HPO_4, anhydrous, reagent grade, 1 gram; trypan blue, 1.0 per cent, 7.5 ml.; crystal violet (Commission Certified), 0.10 per cent based on dye content, 0.8 ml.; water, 1,000 ml. Sterilize as usual. When cool enough to pour, add 0.20 ml. of 5.0 per cent *unheated* "certified" potassium tellurite and pour into 40 plates. Since the amount of tellurite is critical, I have prevailed upon the Difco Laboratories to make, in conjunction with experimental lots of the dehydrated medium, a supply of tellurite solution which I have titrated.

After exactly 24 hours of incubation on this medium at 37° C. (some minute colonies of *Streptococcus mitis* may be more easily distinguishable with longer incubation), *S. salivarius* produces blue smooth or rough "gum drop" colonies from 1 to 5 mm. in diameter which can be easily recognized by reflected light. *S. mitis* produces small blue colonies 0.5 mm. or less in diameter which are slightly raised. Enterococci produce dark blue or black shiny, slightly raised colonies, 1 to 2 mm. in diameter, which have a characteristic appearance and are easily differentiated from *S. salivarius* and *S. mitis*, particularly when viewed by reflected light. Other types of streptococci, which are uncommon, have not been studied. Coliforms may grow occasionally, but they produce brown colonies and rarely spread. The pleuropneumonia group produces mucoid colorless colonies from 5 to 10 mm. and more in diameter. Molds grow after two days.

In vitro *Testing of Toxigenicity of Streptococci.* This test of the toxigenicity of streptococci is based on the facts that (1) toxigenic bacteria resist the antibacterial action of fresh, diluted, defibrinated guinea pig blood, [32-47] and (2) streptococci that resist the antibacterial power of the fresh guinea pig blood also resist appropriate time/concentrations of a number of injurious agents.[16] These relationships have been substantiated by innumerable subsequent experiments in this laboratory and elsewhere.

The method used to test streptococci is as follows:—Cut 10 *S. salivarius* and 10 *S. mitis* colonies from the medium, using an inoculating needle which has been flattened at the tip like a spear. Shake each colony and the attached medium into a 100 x 25 mm. test tube containing 10 ml. of brain heart infusion (Difco). Place the tubes in the incubator. *S. salivarius* begins to produce turbidity or sediment in 6 hours and *S. mitis* may begin to show growth in about 12 hours. After this initial period, take the tubes from the incubator every hour and inspect them for evidence of growth. Remove those that are sufficiently cloudy or have a sufficient amount of sediment, place them in the refrigerator, and test as follows as soon as possible afterwards: If the growth occurs almost entirely as a sediment, decant enough supernatant to make the resulting suspension approximately of standard density, which is easily recognized by the experienced worker. Mix a loopful of the growth with 0.5 ml. of 1:125,000 hexylresorcinol. If the amount of growth is too small, use a drop instead of a loopful of culture. Streak a loopful of the resulting suspension halfway across a blood agar plate to serve as a control of the density of the growth. Make 10 to 12 streaks on

each plate. Immediately incubate the suspensions for exactly 110 minutes, timing them from the start of each plate and then streak a second loopful, making it continuous with the corresponding control streak to permit easy comparison. Discard the tubes and incubate the blood agar plates exactly 24 hours, not "overnight." Observe any hemolytic growths and any evidence of impurity of the growths and compare the density of growths. Those streaks that show as dense a growth in the second streak as in the control are toxigenic.

Distribution of Number and Proportion of Toxigenic Streptococcus salivarius *and* Streptococcus mitis *in Man.* The nasal cavities should contain no toxigenic streptococci, but the properly obtained (*i.e., thoroughly* swabbed, particularly behind posterior pillars and any hyperemic areas) pharyngeal swab contains them in proportion to the extent of local intoxication, which may not, however, coincide with the appearance of the membrane. Normally, a properly obtained throat swab contains less than 200,000 total streptococci and less than 20 per cent of them are toxigenic. *Streptococcus salivarius* and *S. mitis* are present in approximately equal number and proportion in most instances. On the contrary, gingival crevices or pockets and extracted teeth contain practically no *S. salivarius.* In the absence of gingivitis, a gingival swab contains about as many *S. mitis* as does a throat swab of the same person, and the proportion of toxigenic colonies is also similar. Non-infected teeth, smashed on a steril anvil, rarely contain more than 1,000 total streptococci and less than 20 per cent of them are toxigenic. When the dental and periodontal tissues are infected, the number and proportion of toxigenic streptococci rise in proportion to the extent and severity, respectively, of the toxic infection.[5,22,31] Contrary to expectation, the bulk of the dental infection in teeth without peridentitis is in the chamber.[5]

When toxigenic streptococci are swallowed and the gastric acidity is within normal limits, the toxic products inhibit proliferation of *Escherichia* and *Aerobacter* and reduce the lactose fermenting power of these organisms in the gastrointestinal tract.[15,18] However, in the absence of gastric HCl, there is nothing to check the coliforms initially, and they are then able to proliferate rapidly, regardless of the presence of streptococcus toxins.[15,21] This relationship is so constant that the fecal coliform count can be used as an indicator of achlorhydria.

These relationships apply to semisolid specimens obtained after a laxative or from persons with diarrhea. In the formed stool, there is a considerable increase in coliforms and enterococci and a marked decrease in streptococci. In semisolid feces, however, and with normal gastric acidity, the greater the streptococcal intoxication the fewer the coliforms (and the more paracoli). The number of fecal bacteria is best expressed in millions per 100 grams of dry feces. The normal stool contains about 100,000 coliforms, but the toxic stool may contain less than 1,000 and in achlorhydria the number rises above 1,000,000.

The number of streptococci in the semisolid fecal specimen probably represents the ability to multiply in the gastrointestinal tract as a result of being able to overcome local defensive measures, thus making the total number of streptococci a rough index of their pathogenicity, as in other parts of the body. However, in highly toxic stools, streptococci may be inhibited and may even disappear. The proportion of toxigenic variants, on the other hand, represents the severity of the intoxication.

Hematological Considerations. Better understanding of hematological relationships of streptococci in man depends,

above all, on a more accurate and more significant hematological technique than is generally used. The leucocyte count pipette must be mechanically shaken two minutes and two drops must be counted. If they disagree, further counts must be made. The film must be spread rapidly immediately after the drop of blood has been obtained. It must be thin enough so that, in a considerable portion, the leucocytes are well isolated and sufficiently spread out to reveal the finer nuclear structure clearly. The dye solution must be made from a Commission Certified or other satisfactory batch of dye, suitably diluted so as to preserve its staining properties; the proportion between dye and buffer diluent must be 1:2 (by volume, not the number of drops); and the timing must be adjusted to give best results. At least 200 cells must be counted and differentiation of maturity from immaturity made on the basis of the presence or absence, respectively, of a filament, not a narrow band. The nonfilament-filament ratio is expressed in per cent.[23] This technique permits better understanding of hematological relationships, and the normal range is much narrower than the present arbitrary one. Experience in this laboratory indicates that, instead of 5,000 to 10,000 per cu.mm., the normal total leucocyte count is from 7,800 to 8,900; the polymorphonuclears, from 60 to 66 per cent; the mature polymorphonuclears, from 45 to 55 per cent; and the nonfilament-filament ratio, from 15 to 22 per cent. These values appear to be borne out by the effect of leucocyte-stimulating agents,[49] the total number of mature leucocytes being inversely proportional to the amount of leucocyte-stimulating agent required to bring the amounts within the newly established normal limits.

In a few, mostly early cases, the values are above normal due to stimulation of leucopoiesis, but in most persons with low-grade chronic infection the toxins have acted so long that they have had an unfavorable effect on leucopoiesis and the values are subnormal. Usually, the greater the streptococcal intoxication, the lower the hematologic values. The relationship is quite close with streptococci but not with other bacteria.

Some persons show inhibition of some cell types but normal or even stimulated values in others, making it difficult to appraise the results. More recent investigation has shown the extreme importance of the mature polymorphonuclear neutrophilic leucocyte (the "fighter"). The proportion may not always reflect the true degree of toxic effect, but this defect has been largely overcome by calculating the total number of mature leucocytes. While this requires unusual technical skill, the results are well worth it. The total number of mature leucocytes in non-infected persons and those with no toxic inhibition of leucopoiesis is 3,600 per cu.mm., but about 90 per cent of persons with low-grade chronic infection show values below this figure, the depression being proportional to the loss in defense against infection. When the number of total leucocytes is above normal, the total number of mature leucocytes should be proportionately higher or the leucocytosis will be inefficient.

Though the blood sedimentation rate is less constantly related to the number and proportion of toxigenic streptococci, such a relationship actually does exist.[23]

The bacteriological and hematological values follow a seasonal trend with peaks in May and December and troughs in February and August. Likewise, there may be marked differences from the average trend at any particular time, due to inadequately understood climatic, epidemic, and other conditions.[50-52]

Realizing the many shortcomings of the methods, it is possible to allocate findings in different degrees of infection or intoxication, as shown in TABLE 1.

TABLE 1

RELATION OF DEGREES OF INFECTION OR INTOXICATION TO THE DIFFERENT VALUES DISCUSSED

	Relatively non-infected persons	Mild infection or intoxication	Moderately severe infection or intoxication	Severe infection or intoxication
Total number of streptococci:				
Pharynx	Less than 200,000	200,000 to 1,000,000	1,000,000 to 20,000,000	Over 20,000,000
Gingivae (*S. mitis*)	Same as the pharynx	2 to 3 times the pharynx	4 to 10 times the pharynx	More than 10 times the pharynx
Extracted teeth (*S. mitis*)	1,000	1,000 to 50,000	50,000 to 500,000	Over 500,000
Feces: intox.	Over 1,000	200 to 1,000	50 to 200	Less than 50
infection	Less than 50	50 to 200	200 to 1,000	Over 1,000
Proportion of toxigenic streptococci:				
Pharynx	Less than 20%	25 to 50%	50 to 80%	Over 80%
Gingivae (*S. mitis*)	Same as the pharynx	10% more than the pharynx	20% more than the pharynx	Over 30% more than the pharynx
Extracted teeth (*S. mitis*)	Less than 20%	25 to 50%	50 to 80%	Over 80%
Feces	Less than 25%	25 to 50%	50 to 80%	Over 80%
Coliforms in feces:				
In achlorhydria	100,000	The number is above 1,000,000, may rise to 20,000,000		
Otherwise	100,000	40,000 to 90,000	5,000 to 40,000	Less than 5,000
Paracoli	None	Number usually increases with intoxication giving a high paracoli:coli ratio.		
Sedimentation rate (Westergren), mm in 1 hour (fresh blood):	Less than 5	5 to 20	20 to 50	Over 50
Stimulation of leucocytes:				
Total WBC	7,500-8,900	9,000-10,000	10,000-12,000	Over 12,000
Nonfil.-fil. ratio	20%	20-30%	30-75%	Over 80%
Polys.	60-66%	68-70%	70-75%	Over 75%
Mature polys.	45-55%	These are rarely over 55%, even in acute infections		
		The higher the percentage, the more efficient the leucocytosis		
Monocytes	2-4%	4-5%	5-7%	Over 7%
Eosinophiles	2-3%	3-4%. Higher values are more characteristic of allergy		
Depression of leucocytes:				
Total WBC	7,500-8,900	6,000-7,500	4,500-6,000	Below 4,500
Nonfil.-fil. ratio	20%	Values usually increased, but often rise with effective response, indicating that apparently low values were actually depressed values.		Below 20%
Polys.	60-66%	52-60%	45-52%	Below 45%
Mature polys.	45-55%	40-45%	30-40%	Below 30%
Total mature polys.	3,600	2,500-3,500	1,800-2,400	Below 1,800
Monocytes	2-4%	2%	1-1.5%	Below 1%
Eosinophiles	2-3%	1%	0.5%	None
Bacterial products in urine (units):	Less than 5	6 to 15	16 to 30	Over 30

Lab. number
Patient
Physician

Date begun.....

		Line	"Normal" range[1]	Findings in the average chronic invalid[2] Low	Medium	High
HEMATOLOGY						
Leucocytes: Total	per cu. mm.	1	7,500 to 8,800	5,900	7,600	9,500
Total mature	per cu. mm.					
Neutrophiles,						
filamented (mature)	%	2	50 to 6		34	
nonfilamented	%	3	7		20	
total	%	4	60 to 65	45	54	62
nonfilament-filament ratio	%	5	15[3]	40	60	91
Lymphocytes	%	6	30 to 40		38	
Monocytes	%	7	2 to 4	3.4	5.1	7.6
Eosinophiles	%	8	2 to 3	1.0	2.4	4.5
Basophiles	%	9	0.5	0.3	0.7	1.3
Blood sedimentation rate (Westergren)	mm. in 1 hour	10	less than 5	9	20	39

Microorganism[9]	Total number[4,5]	Percentage of toxigenic colonies[6]
BACTERIOLOGY		
NASAL CAVITIES		
Streptococcus salivarius[7]	11 none	
mitis	12 none	
hemolyticus	13 none	
Pathogenic staphylococci[8]	14 none	
Other bacteria	15 none	
THROAT		
Streptococcus salivarius	16 less than 250,000 and	40,000,000 and 67%
mitis	17 less than 20% of each	25,000000 and 62%
hemolyticus	18 none	
Pathogenic staphylococci	19 none	
Other bacteria	20 none	
GINGIVAL CREVICES OR POCKETS		
Streptococcus salivarius	21 less than 50,000 and less than 20%	50,000 and 18%
mitis	22 less than 250,000 and than 20%	same as in the pharynx of the person
hemolyticus	23 none	
Pathogenic staphylococci	24 none	
Other bacteria	25 none	
TOOTH		
Streptococcus salivarius	26 none	
mitis	27 less than 1,000 and less than 20%	
hemolyticus	28 none	
Pathogenic staphylococci	29 none	
Other bacteria	30 none	
FECES		
Escherichia (*B. coli*) and *Aerobacter*	36 75,000 to 250,000	
Paracoli (lactose nonfermenters)	37 none	
Enterococci	38	
Streptococcus salivarius	39 none	
mitis	40 none	
hemolyticus	41 none	
Pathogenic staphylococci	42 none	

REMARKS

Low counts indicate that toxins are preventing leucocytic response, which is characteristic of severe intoxication and low resistance, whereas high counts indicate active response to the infectious agent. Since these are the "fighters," the percentage is proportionate to the defense against infection.

Same as line 1.
Except in severe infections with poor resistance, which produce a low ratio, the ratio generally is proportionate to the severity of the infection.

Same as line 1.
Same as line 1. Eosinophilia may be associated with hypersensitivity to the infection.

Usually, the rate is proportionate to the severity, extent and duration of the infection.

Nasal streptococci and staphylococci may be caused by diffuse inflammation of the lining membranes or may be draining from the sinuses.

Same as line 20.

The higher the number of streptococci the more sure and more extensive the infection. The higher the percentage of toxigenic colonies, the more severe the intoxication.

Pathogenic staphylococci have local rather than general significance.
Other bacteria, usually coliforms, occur when the infection is severe.

This occurs in small numbers in the gingival margins, but may increase when local resistance is low.
Normally, the number and percentage of *S. mitis* are similar to those in the throat. The higher the gingival counts, the surer and more severe the gingivitis.
Rarely found in gingival crevices.
Same as line 20.
Same as line 20.

This is uncommonly found in aseptically extracted teeth.
The higher the number and percentage of toxigenic *S. mitis*, the more severe and more extensive the intoxication in the tooth. Teeth with counts above 1,000,000 are likely to be major foci of infection.
Hemolytic streptococci are rare in teeth.
Same as line 20.
Same as line 20.

In achlorhydria, the count is above 1 million. Otherwise, the lower the count, the more severe the intoxication.
In severe intoxications, there may be almost complete absence of all types of bacteria.
Paracoli are characteristic of severe intoxication. Usually, the higher the number the more severe it is.
Enterococci are nonpathogenic in the majority of cases.
The percentage of toxigenic streptococci is proportionate to the severity of the intoxication. When the intoxication is caused by swallowed toxins, there may be few streptococci in the feces. An increase in the fecal streptococci suggests actual invasion of the intestinal mucosa.
Usually residual from the upper respiratory tract, but often contribute to intestinal symptoms.

CHART 1 (*continued*).

Excreted Bacterial Products. The author has devised a method of titrating certain bacterial products in urine that takes about 30 seconds and requires only 1.0 ml. of sample. To do the test, add 10 per cent Roccal drop by drop to 1.0 ml. of urine until the precipitate which increases and then decreases in turbidity shows no further decrease. The end-point is an opalescence in most cases. Each 0.01 ml. of reagent required is considered equivalent to 1 unit of bacterial products. Persons with little bacteriological and hematological evidence of infection show less than 5 units, but in severe infections the value may rise above 100.

Theoretically, when streptococci multiply in the relatively immune person, the metabolic products are rendered innocuous by the defense mechanism, but, when the effectiveness of the defense mechanism has been reduced, some of the bacterial products are unaffected and are excreted unchanged in the urine. This hypothesis is confirmed by the new titration method. When the mature leucocyte count is normal or above normal (*i.e.*, 3,600 or more per cu.mm.), the amount of titrated bacterial products in the urine is low, whereas when the number of mature leucocytes is low the amount of bacterial products in the urine is much higher, usually in proportion to the extent of depression of the total number of mature leucocytes. Hence, the titer appears to indicate the amount of bacterial products that the body has been unable to destroy and, if so, is another measure of the defense mechanism. The method has been used successfully in following response to treatment of the infection.

This method is suggested here (as a preliminary announcement) only because of its connection with the other subjects discussed and because it is hoped that it will stimulate others to study what appears to be a rapid, simple method easily adaptable to bedside technique. At present, no claim is made as to the type of bacterial product which it measures, nor as to the specificity. These phases, along with study of other high molecule agents of a similar type, will be discussed elsewhere.

Explanation of superscripts in CHART 1 (*see pages* 50-51):

1. Theoretically, a normal person is free from infection, but this rarely occurs and the "normal" person is usually considered one without severe infection. The normal values here, however, are those of persons who have negligible infection.
2. The bacteriology, hematology, and severity of infection are subject to considerable seasonal variation. Usually, the values are comparatively mild in February and August and severe in May and December, but they may differ from this pattern.
3. Although the "normal" nonfilament-filament ratio (the Schilling index) is considered to be 6 per cent, very few people show such a low value. Our average in relatively non-infected persons is about 20 per cent and values below this indicate that the toxins have prevented the leucopoietic tissues from responding to the toxic stimuli.
4. Special methods have been developed in this laboratory for isolating pathogenic types of staphylococci and streptococci. They permit reliable isolation even when numerous other bacteria are present. Hence, the proportion of positive findings is much higher than elsewhere.
5. Fecal bacteria are counted on the basis of millions per 100 grams of dry feces. Other foci are estimated on the basis of the total number in the swab or tooth. Teeth are broken aseptically. Experiments show conclusively that in carious teeth the bulk of the intoxication originates in the streptococci in the cavity, even when the x-ray indicates periapical involvement. In such cases, there are very few toxigenic streptococci around the apex of the tooth, but large numbers in the chamber. When there is extensive involvement of the root canal, the root fragments also show a high count. Non-vital teeth usually show higher counts than do vital teeth, but since the number and percentage of toxigenic streptococci is proportionate to the extent and severity of decay, the relation to vitality is only a general one.
6. Toxigenic streptococci, the common denominator of chronic illness, produce a toxic principle which is distinct from other pathogenic factors and may be readily demonstrated by an appropriate technique. About 20 colonies of streptococci are tested from each sample. The percentage of toxigenic colonies usually is proportional to the severity of the infection in the part tested. These toxigenic streptococci make excellent antigens, whereas nontoxic strains have little value. The resistance of streptococci to certain injurious agents has been shown to be parallel with the toxigenicity of the cultures. Its precise determination depends upon careful attention to numerous technical details.
7. The old classification of nonhemolytic streptococci as "*viridans*" and "nonhemolytic" is inaccurate. The differentiation is now based on careful and extensive study of biochemical properties. Generally, "nonhemolytic" streptococci are *Streptococcus salivarius* and "*Streptococcus viridans*" are *Streptococcus mitis*.
8. Staphylococci that coagulate blood are probably pathogenic. Others are ignored except in special cases.
9. Most non-pathogenic bacteria or those of little pathologic significance are ignored.

Summary of Research Project: Distribution, extent, and toxicity of streptococci and distribution of pathogenic type staphylococci in possible foci of infection.

Date of report
Patient
Physician

HEMATOLOGIC FINDINGS

	Stimulated response (degree above normal)	Toxic inhibition (degree below normal)
Nonfilament-filament ratio		
Total leucocytes		
Neutrophiles		
Mature neutrophiles, per cent		
Total mature neutrophiles		
Monocytes		
Eosinophiles		
Acceleration of blood sedimentation rate		
Urine polysaccharide titer		

BACTERIOLOGIC FINDINGS

	Streptococcal toxin			Pathogenic type staphylococci
	Extent (based on relative number)	Intensity (based on relative percentage of toxigenic)	Intensity (based on coliforms)	
Nose				
Throat				
Feces				

CHART 2.

Reporting the Results of the Tests. Although numerous different report forms have been worked out from time to time, it is difficult to include all necessary data and retain simplicity. More recently, the detailed report form (CHART 1) has been accompanied by a Summary of Research Project form (CHART 2). The forms are reproduced here, therefore, not with any dogmatic finality but with the idea of pointing the way to clearer reporting of the different findings.

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Note. The Section of Psychology held a meeting on November 17, 1947, at which Doctor Gustave Mark Gilbert, Metropolitan State Hospital, Waltham, Mass., presented a paper entitled, *An Analysis of the Psychological Examinations Administered to the Nazi Leaders at Nuremberg.* No abstract of this has been received.

The Section of Anthropology held a meeting on November 24, 1947, at which Mr. Harry Tschopik, American Museum of Natural History, New York, N. Y., presented a paper entitled, *Chucuito: an Aymara Indian Community in Southern Peru,* illustrated by motion pictures and recorded music. This will be published in a later issue of the present volume of TRANSACTIONS.

SECTION OF PHYSICS AND CHEMISTRY

ADSORPTION AS A RESEARCH TOOL

By A. C. ZETTLEMOYER*

During the past ten years, extensive developments have been made in the application of gas adsorption techniques to the solution of research problems. This period began with the work of Brunauer, Emmett, and Teller, who for the first time devised a method for determining the surface area of a solid with considerable precision and accuracy. At the present time, the physical and chemical structure of the surface can often be elucidated by gas adsorption studies. If the solid material is non-porous, average particle size can also be determined. Due to these developments and to the importance of a knowledge of the properties of interfaces in many technological developments, many companies in recent years have added gas adsorption research groups to their research laboratories. Today, gas adsorption techniques are used to study adsorbents, catalysts, pigments and, indeed, any solid for which interaction with gases or liquids is the important phenomenon.

One of three types of gas adsorption apparatus is usually employed. The Emmett and Brunauer apparatus is the most common. For surface area measurements, nitrogen is used as the adsorbate and the adsorbent is held at the temperature of liquid nitrogen. The nitrogen gas is introduced from a burette through a dead space kept as small as possible to minimize the dead space correction. Helium is used to calibrate the dead space; this gas is not adsorbed to any appreciable amount at the temperature of liquid nitrogen. The temperature of the bath is usually measured with a nitrogen vapor pressure thermometer. This type of apparatus possesses the disadvantage that the equilibrium points are approached from pressures above the equilibrium pressure going up the isotherm and from pressures below going down. Nitrogen is used because it is a small molecule, is inert, and may be readily obtained in a pure state.

The McBain-Baker balance is used for studying the adsorption of organic vapors and for high-temperature adsorption studies. This apparatus makes use of a quartz spiral spring to hold the small bucket containing the sample. As the weight of the sample increases due to adsorption, the extension of the spring is followed with a cathetometer. No dead space correction is necessary. An interesting modification of this apparatus, using a beam instead of a spring, has recently been devised by Gregg.

The Wooten-Brown apparatus is used for the measurement of solids having areas below one square meter per gram. For such low areas, the amount adsorbed will be relatively small. Therefore, the dead space correction is greatly reduced by using a gas far below its condensation pressure; for example, krypton may be used at the temperature of liquid nitrogen. Then, even at high relative pressures, very little gas will be in the dead space. Mercury cutoffs must be used instead of stopcocks and pressures are read on a McLeod gauge.

A number of interesting problems with regard to pigments, adsorbents, and chemical reagents have been solved in the Lehigh Gas Adsorption Laboratory. An especially interesting one concerns yellow iron oxide. When this monohydrate is prepared by the corrosion process in the presence of certain impurities, a richer-looking product is produced than when these impurities are not present. This pigment forms a transparent coat-

* William H. Chandler Chemistry Laboratory, Lehigh University, Bethlehem, Pa. This paper, illustrated by lantern slides, was presented at the meeting of the Section on November 28, 1947.

ing when dispersed in linseed oil. While usual physical tests such as x-ray failed to distinguish the new variety from the old, differences could easily be distinguished by gas adsorption technique. The surface area of the new type was found to be about ten times greater than that of the older type and the pore size of the new was found to average only one-half that of the old.

Another problem involved a series of nine manganese dioxide ores which were examined for differences in surface area. These materials showed a wide difference in activity when used in dry cells. The surface areas differed in the same order. Thus, a technique for predicting their activity was worked out. Activity, of course, is not necessarily dependent on surface area but depends also on the nature of the surface. It is activity per unit area which is the more basic factor. In the case of the dioxide samples, as often happens, the extent of the surface is the important factor.

Recently, we have concerned ourselves with adsorption on organic surfaces. This field has been little explored but offers excellent possibilities for the use of the adsorption technique. The work of Pauling on the structure of proteins, for example, has indicated that the surface structure is the controlling factor in deciding the specificity of the activity of the protein. While some work has been done in our laboratory on gas adsorption on collagen, it appears necessary to initiate a systematic study of adsorption on organic surfaces. This must necessarily begin with studies of physical adsorption on pure hydrocarbon surfaces. This work has just gotten under way.

Much of the work in the Lehigh Adsorption Laboratory has been and is concerned with Active Magnesia, work done under the auspices of the Westvaco Chlorine Products Corporation. We started work on this material several years ago when it was used in making 1707 catalyst for the synthetic rubber program. Low-temperature nitrogen adsorption isotherms were measured on a series of commercial active magnesias. The isotherms were mixtures of types II and IV. The isotherm shape was explained by hypothesizing a structure consisting of plates of checkerwork of cubic holes and crystallites. The surface areas of the magnesias were shown to range from less than 1 to more than 200 square meters per gram. Some difficulty was encountered in the use of the usual methods for calculating areas from adsorption data. The areas were shown to be directly proportional to the amount of iodine adsorbed from solution, or the iodine number.

Active Magnesia is now used as an adsorbent in removing silica from boiler water, fatty acids from petroleum ether, and impurities from petroleum lubricants, dry cleaning solvents and other non-aqueous and unreactive fluids, and in chromatography. Active Magnesia shows good promise also as a basic, solid catalyst for vapor phase organic reactions.

Because the BET plots for the active magnesias were always found to be slightly concave to the pressure axis, there was some concern as to whether the BET theory could actually be applied. Modifications of the BET theory which were in the literature were investigated without success. A dual surface adsorption theory based on the multi-molecular theory of Brunauer, Emmett, and Teller was found to explain adequately the adsorption isotherms. This theory involves the concept that commercial magnesium surfaces are made up of two portions differing in their heats of adsorption. The one portion consisting of about 25 per cent of surface has a very low heat of adsorption for nitrogen, and this means that the c value connected to the BET theory is very low for this portion.

Since magnesium oxide has a regular cubic lattice made up of magnesium and

oxide, it is very adaptable to a precise study of adsorption. The 100 face of the crystal is the one most always found. Therefore, a number of precise studies have been made of the physical adsorption of gases on pure magnesium oxide.

Physical adsorption of gases on solids is generally considered to take place with a close-packed first layer. Using this concept, the area occupied per molecule can be calculated from the density of the normal condensed phase. Areas calculated accordingly from the adsorption of various adsorbates on active magnesias were non-concordant.

To explain these discrepancies, the implications of adsorption in preferred positions were examined. From the geometry of the 100 face of a magnesium oxide crystal, distances between preferred positions were determined. Eigenvalues for the area per adsorbed molecule could then be calculated as a function of these distances and the size of adsorbate molecules. These eigenvalues for the effective molecular areas are simple multiples of a constant characteristic of the crystal face. This relationship leads to the conclusion that, when adsorption takes place in preferred positions, the ratio of the moles of adsorbate A to the moles of adsorbate B required to complete a unimolecular layer on the surface can be expressed in terms of simple whole numbers.

Data for the adsorption of a number of gases and for adsorption of iodine from solution on chemically proven magnesium oxide were found to be in excellent agreement with this postulate.

NEW MEMBERS

Elected November 20, 1947

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SUSTAINING MEMBERSHIP

Whymper, Robert. Food Consultant, New York, N. Y.

ACTIVE MEMBERSHIP

Adams, Frederick W., D.Sc. Director of Research, Clark Thread Co., New York, N. Y.

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